

Modeling of long term behavior of ablation plumes produced with ultrashort laser pulses

A.M. Komashko, M.D. Feit, A.M. Rubenchik

This article was submitted to
Photonics West 200 Conference Proceedings,
San Jose, CA, January 22-24, 2000

U.S. Department of Energy

Lawrence
Livermore
National
Laboratory

February 10, 2000

DISCLAIMER

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

This is a preprint of a paper intended for publication in a journal or proceedings. Since changes may be made before publication, this preprint is made available with the understanding that it will not be cited or reproduced without the permission of the author.

This report has been reproduced
directly from the best available copy.

Available to DOE and DOE contractors from the
Office of Scientific and Technical Information
P.O. Box 62, Oak Ridge, TN 37831
Prices available from (423) 576-8401
<http://apollo.osti.gov/bridge/>

Available to the public from the
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Rd.,
Springfield, VA 22161
<http://www.ntis.gov/>

OR

Lawrence Livermore National Laboratory
Technical Information Department's Digital Library
<http://www.llnl.gov/tid/Library.html>

Modeling of long term behavior of ablation plumes produced with ultrashort laser pulses

Aleksey M. Komashko*, Michael D. Feit, Alexander M. Rubenchik

Lawrence Livermore National Laboratory, P.O. Box 808, mail stop L-438

Livermore, CA 94550 USA

ABSTRACT

Expansion of ablation plumes created by intense ultrashort lasers is determined by various complicated physical processes which have very different spatial and time scales. Since complete simulation by one model is practically impossible, we suggest using two models describing initial and final stages that can be matched at an intermediate time. The proposed modeling procedure connects laser parameters to plume properties far away from the ablation spot. Laser material interaction and beginning of the expansion are simulated with a one-dimensional hydrodynamics code and the final stage is modeled using an analytical solution for an expanding three-dimensional ellipsoidal gas cloud.

Keywords: Simulation, ultrashort laser pulses, ablation, expansion, ellipsoid model, plume directionality

1. INTRODUCTION

For a number of experiments involving the interaction of intense ultrashort laser pulses with matter, it is desirable to know directionality, velocity and other parameters of ejecta far from the target. Laser film deposition¹, laser breakdown diagnostics² and material processing³ can be viewed as typical applications. A variety of physical processes, for instance laser-material interaction, hydrodynamics, thermal and radiative transport, make analytical description extremely difficult. In this case numerical simulations are the only viable option. However, the large disparity in spatial and temporal scales involved often makes it impractical to simulate both ablation and ejecta expansion to large distances from the target within the framework of one model. For example, a typical calculation would require an adequate description of laser energy deposition which occurs on a time scale of hundreds of femtoseconds and on a spatial scale on the order of tens of nanometers. Plume modeling has to predict behavior of the ejecta at the final stage of expansion that takes place microseconds and centimeters away from the initial moment.

A possible solution to this problem is to employ more than one model, each suitable for a particular stage, and match them at an intermediate time. Plume evolution includes two important parts – the initial stage is one dimensional (laser spot large compared to skin depth) and the final consists of three-dimensional expansion. The difference is not only geometrical, but in the physics playing the major, defining role. The initial one-dimensional stage occurs during laser energy deposition and the initial expansion takes place. The longitudinal size of the hot material is still much smaller than the transverse size determined by the laser spot. Clearly, laser-material interaction, ionization, energy and material transport are the significant effects during this period. After some time, the ejecta cloud becomes truly three-dimensional, but the physics simplifies considerably. At this stage, plume evolution is described by conventional hydrodynamics and is sensitive only to integral parameters of the laser pulse.

* Correspondence: Email: komashko1@llnl.gov, Telephone: +1-925/422-4754, Fax: +1-925/422-5537

Modeling of the first stage connects laser and target parameters with ejecta description. The integral features of ejecta determine three-dimensional plume expansion. In this paper, we used the one-dimensional radiation-hydrodynamics code HYADES⁴ with laser package to simulate the initial stage and modeled the full geometry expansion using an idea suggested by Anisimov et.al.⁵ based on an exact analytical solution for an adiabatically expanding gas ellipsoid. We propose matching criteria and discuss long term plume behavior as a function of laser parameters.

2. THREE-DIMENSIONAL MODEL

At long times, the ablation plume is described by ordinary hydrodynamics and should be sensitive only to integral properties of the laser pulse such as the fluence. Anisimov et.al.⁵ suggested an exact hydrodynamic self-similar solution for 3D gas ellipsoid expansion to describe such ejecta. In this model, the plasma remaining after pulse termination is modeled as an ellipsoid with three different axes and expands adiabatically outward. All flow parameters are constant on ellipsoidal surfaces for this solution. The description of the process of expansion includes gas cooling and cloud shape deformation. However, this deformation only changes the ellipticity, the cloud remains an ellipsoid.

In general, the hydrodynamic equations describing gas cloud expansion are difficult to solve, but under certain assumptions, these partial differential equations can be reduced to ordinary differential equations which greatly simplifies our problem. This transition is based on the assumption that we have a self-similar solution, i.e. that every physical parameter distribution preserves its shape during expansion. Then, separation of variables, namely time and Lagrange space coordinates is possible. These types of coordinates are widely used in hydrodynamics because they simplify the equations. Very often Lagrange space coordinates are initial coordinates of the particles. Here we follow this choice.

It has been shown⁶ that for an adiabatically expanding ideal gas ellipsoid all physical parameters are constants on ellipsoidal surfaces evolving in time. In this case, the dynamics of the plume is defined by the ordinary differential equations describing evolution of the ellipsoid axes:

$$X \frac{d^2 X}{d\tau^2} = Y \frac{d^2 Y}{d\tau^2} = Z \frac{d^2 Z}{d\tau^2} = \left(\frac{A}{XYZ} \right)^{\gamma-1}, \quad (1)$$

where τ is dimensionless time, γ is the adiabatic constant and A is a function of initial plume size. The dimensionless functions X, Y, Z define the ellipsoidal shape at any particular time. Eq. (1) can be easily solved numerically. Once the functions X, Y, Z are known, all thermodynamical plume parameters can be calculated. Self-similar solutions exist in two different situations ; expansion can be isentropic or isothermal. Isentropic expansion has a well defined plume size and parabolic profiles of physical parameters. Isothermal expansion results in a Gaussian density profile. The choice between these two options should be made in reference to the one-dimensional modeling. However, geometrical properties of the plume, for example directionality, are completely independent of this choice.

Note that Eqs. (1) can be viewed as equations of motion in a repulsive potential

$$U(X, Y, Z) = \frac{1}{\gamma-1} \left(\frac{A}{XYZ} \right)^{\gamma-1}$$

with conservation of “energy”

$$e = \text{const} = \frac{1}{2} (\dot{X}^2 + \dot{Y}^2 + \dot{Z}^2) + U(X, Y, Z).$$

This is a mathematical representation of the fact that after the initial stage, the plume begins to expand inertially. Thermal energy is transferred into kinetic energy and finally all particles fly away at constant velocity. The direction and particular value of this velocity depend on initial parameters of the plume and γ . We will assume that the adiabatic index equals 5/3.

Numerical integration of Eqs. (1) shows that the plume very quickly reaches the inertial stage. By the time it grows to centimeter size, its shape is already frozen. To characterize the final stage we introduce the ratios $k_y \approx Y(\tau)/X(\tau)$, $k_z = Z(\tau)/X(\tau)$. These functions approach limiting values that define the

leads to a linear relationship between the fields in one layer to the next. That is, they are connected by a 2×2 matrix. The overall properties (transmission and reflection) of the medium are found by multiplying all these matrices together. The fields penetrating the medium and the resultant energy deposition (Joule heating) can then be simply found. The advantage to this approach is its arithmetical simplicity - it is ideally suited for numerical computation since it involves many simple operations. Effects of angle of incidence and polarization are automatically included. Ability to do this kind of simulation becomes even more important if there is need to take strong prepulses into account.

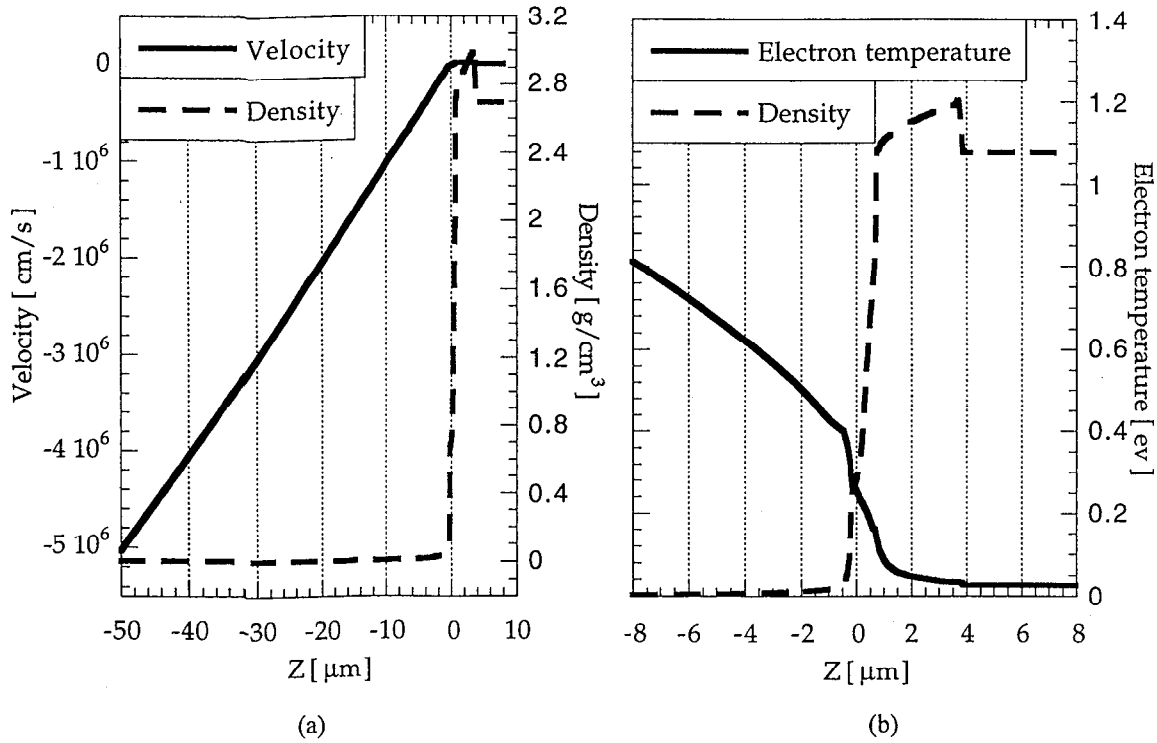


Figure 1. Results of typical HYADES simulation. Material is aluminum. Laser parameters : $\lambda=826\text{nm}$, $\tau=150\text{fs}$, fluence= 12.5J/cm^2 , $\theta=45^\circ$, S polarization. Picture (a) is plot of density and material velocity, picture (b) is density and electron temperature. Both snapshot are at 1 ns.

In a typical HYADES calculation energy absorbed from the laser pulse immediately heats and ionizes material that begins to show some expansion even before the termination of the pulse. Significant deposited power launches a high pressure shock wave into the material; at the same time ablation products expand into the vacuum. If such a calculation is continued for many picoseconds, typical physical parameters will look like those presented in the Fig.(1). The figure shows profiles of density, velocity and temperature after 1ns. From Fig.(1a), one sees that the velocity is a linear function of distance Z. This is an essential feature of self-similar solutions. Other parameters do not have the exact shape given by the analytical ellipsoid solution, however the general behavior is similar to that of the isothermal expansion.

3. MATCHING AND RESULTS

Before one can proceed to modeling, matching criteria have to be defined. Although there is no absolute match in the shape of physical parameters, very often self-similar solutions describe correct asymptotic behavior, even if the initial conditions are not exactly the same. This type of solution and, in particular, the expanding gas ellipsoid solution is governed by integral parameters. If there is a way to identify them from the one-dimensional modeling, matching is possible. That means we have to find the plume mass, energy and initial shape. There is a total of five unknown quantities. The obvious choice for transverse dimensions

are given by the laser spot size. Determining the remaining quantities is not as straightforward. The problem stems from the fact that in the hydrocode simulations there is no plume as such.

A naïve definition of the plume is the mass of material that comes off the target. Clearly mass removal can continue for a long time. We need to devise a method that determines the answer from one-dimensional calculations. Our suggestion is to use a “long time” zero velocity point. Many different runs showed that after a couple of hundred picoseconds the zero velocity point approaches a constant value. Once we decide where the boundary of the plume is, calculation of plume energy and mass is easy. The depth of removed material can be used as the initial thickness.

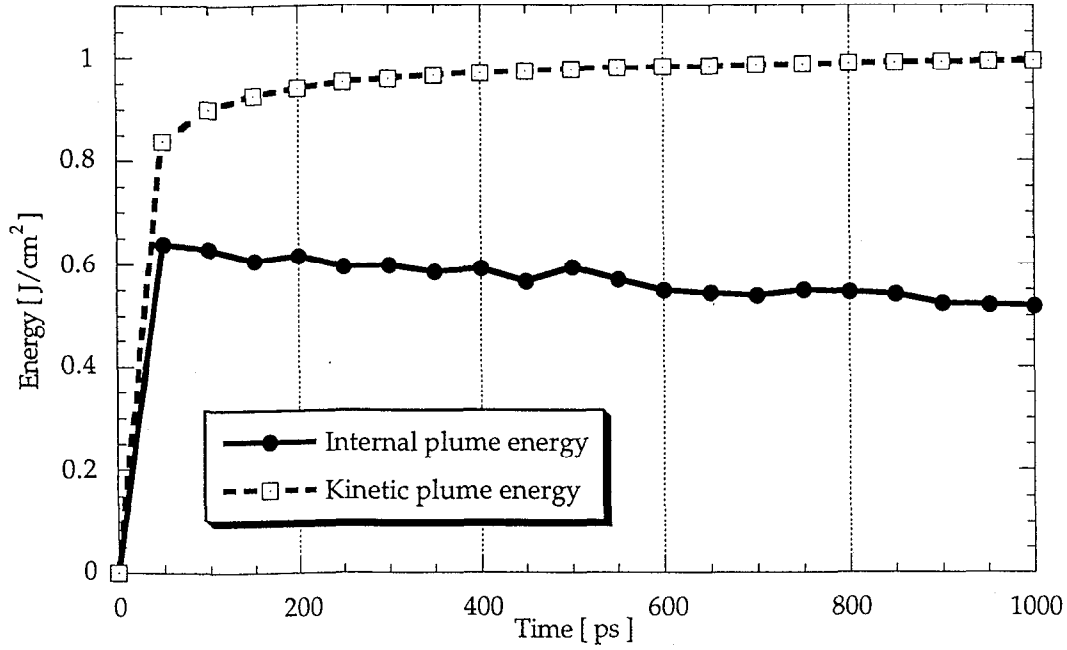


Figure 2. Evolution of the internal and kinetic plume energy. Material is aluminum. Laser parameters : $\lambda=826\text{nm}$, $\tau=150\text{fs}$, fluence= 12.5J/cm^2 , $\theta=45^\circ$, S polarization.

In Fig.(2) we show evolution of the plume energy for a typical run calculated employing this method. The picture demonstrates that indeed there is fast plume formation with a well determined energy. After the plume is created, the effect of initial expansion is evident.

The proposed modeling procedure gives long term plume behavior as a function of laser parameters such as fluence, pulse length, wavelength, etc.. However, the amount of absorbed energy is of utmost significance. Laser energy is deposited in a layer much smaller then the ablated depth; therefore, specifics of the absorption are not very important. For clarity, we present simulations in which only the laser pulse energy was varied. Influence on absorption of other parameters is discussed elsewhere⁸.

Fig.(3) shows several important parameters as a function of fluence: absorbed and plume energy as fraction of incident energy and initial plume thickness. Calculations are for aluminum, which is a highly reflective material. When the pulse energy is high enough to generate a plasma, absorption goes up, reaches a maximum and then decreases because of the plasma mirror effect (at high fluences laser-generated plasma is heated to significant temperatures and the electron collision frequency decreases, thus reducing absorption). Energy in the plume follows the absorption curve pretty closely. This is a demonstration of an important feature of ultrashort laser-material interaction – a large fraction of the pulse energy goes into ablation and its products and only a small part is left in the bulk of the material. The initial plume thickness grows with fluence, but stays below one micron. From Table 1 we can see that even for a hundred micron

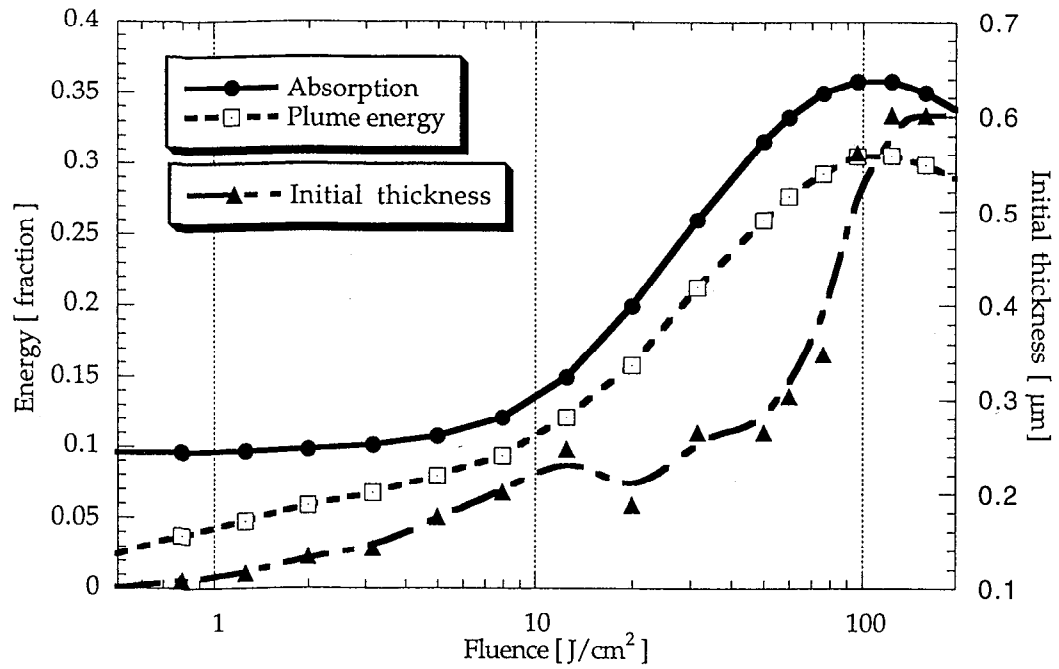


Figure 3. Fraction of energy absorbed, fraction of incident energy in the plume and initial longitudinal plume size as a function of laser pulse fluence. Material is aluminum. Laser parameters : $\lambda=826\text{nm}$, $\tau=150\text{fs}$, $\theta=45^\circ$, S polarization.

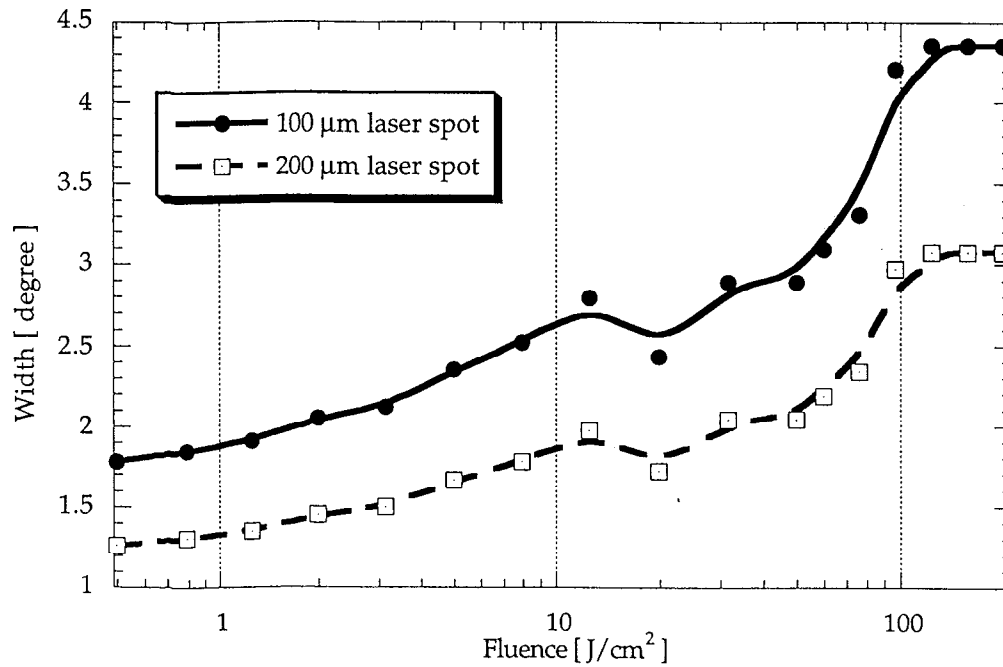


Figure 4. Plume width (hmfw) as a function of fluence for two laser spots. Material is aluminum. Laser parameters : $\lambda=826\text{nm}$, $\tau=150\text{fs}$, $\theta=45^\circ$, S polarization.

laser spot size, plume ejecta will be very directional. Larger spots will only emphasize this effect. Using Table 1 and Eq.(3) we calculated plume directionality as a function of fluence. For simplicity, a circular laser spot is assumed. In this case Eq.(3) simplifies to

$$h(\theta) = H_0(1 + k^2 \tan^2 \theta)^{-\frac{3}{2}}. \quad (4)$$

For convenience index z is omitted. The plume angular half width is given by $\theta_{1/2} \approx 2\arctan(0.56/k)$. Interpolation of numerical results presented in Table 1 gives k as a function of ellipticity a which is the ratio of initial plume thickness to laser spot size. The data can be fitted surprisingly well by a $a^{-1/2}$ function. Since this parameter is small, the formula can be simplified by expanding the inverse tangent. As a result, $\theta_{1/2} \propto \sqrt{a}$. Fig.(4) shows calculation of the plume angular width for two sizes of laser spot and the same one-dimensional calculations as in Fig.(3). Angular size is small and doesn't change significantly with fluence. It is interesting to note that film thickness $H_0 \propto Mk^2$ is also a weak function of fluence – higher fluence means more material is ablated but the plume is less directional, lower fluence gives better directionality but a lower amount of ablated material.

4. CONCLUSION

Two types of models were used to connect ultrashort laser pulse parameters and ablation plume properties far away from the target laser spot. Laser energy deposition and initial expansion were modeled using a one-dimensional hydrocode with laser package. The final stage of plume expansion was treated as an adiabatically expanding gas ellipsoid for which there is an analytical solution. Model matching is implemented through calculation of integral plume parameters such as ablated mass and energy. This modeling procedure predicts scaling of ejecta properties as a function of laser parameters.

ACKNOWLEDGMENTS

This work was performed under the auspices of the U.S. Department of Energy by University of California Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48

REFERENCES

1. D. Bäuerle, "Laser-induced formation and surface processing of high-temperature of superconductors", Appl. Phys. A 48 (1989) 522; L.D. Laude, Ed., Excimer Lasers, NATO ASI Series, Vol. E256, Kluwer, Dordrecht, 1994.
2. T.R. Loree, L.J. Radziemski, "Laser-induced breakdown spectroscopy: a technique for atomic detection and molecular identification", Proc. of the SPIE, 288(1981),241-4; R. Nordstrom, Lasers and Optonics, 23, 1993.
3. M.D. Perry, G. Mourou, "Terawatt to petawatt subpicosecond lasers", Science 264(1994) 917.
4. J. T. Larsen, "HYADES - A Radiation Hydrodynamics Code for Dense Plasma Studies," in *Radiative Properties of Hot Dense Matter*, edited by W.Goldstein, C. Hooper, J. Gauthier, J. Seely and R. Lee, World Scientific, Singapore, 1991; J T. Larsen and S.M. Lane, "Plasma hydrodynamics code for dense plasma studies", J. Quant. Spectr. and Rad. Trans. 51(1994), p.179-86.
5. S.I. Anisimov, B.S. Luk'yanchuk and A. Luches, "An analytical model for three-dimensional laser plume expansion into vacuum in hydrodynamic regime", Applied Surface Science 96-98 (1996), pp. 24-32.
6. I.V. Nemchinov, "Expansion of a tri-axial gas ellipsoid in a regular behavior", Prikl. Mat. Mekh. 29 (1965)1, 134-140.
7. M.Born, E.Wolf, *Principles of Optics*, Pergamon Press, (Oxford 1980).
8. A.M. Komashko, M.D. Feit, A.M. Rubenchik, M.D. Perry, P.S. Banks, "Simulation of material removal efficiency with ultrashort laser pulses", Appl. Phys. A 69(1999) 7, S95-S98.